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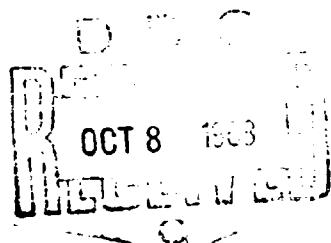
MEMORANDUM REPORT NO. 1934

RADIOLOGICAL HAZARDS OF TRITIUM AND
PROMETHIUM-147 ACTIVATED LUMINOUS DEVICES

by

Richard H. Comer
John D. Knapton

September 1968



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SEPTEMBER 1968

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PROMETHIUM-147 ACTIVATED LUMINOUS DEVICES

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Interior Ballistics Laboratory

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BALLISTIC RESEARCH LABORATORIES

MEMORANDUM REPORT NO. 1934

RHComer/JDKnapton/nlg
Aberdeen Proving Ground, Md.
September 1968

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PROMETHIUM-147 ACTIVATED LUMINOUS DEVICES

ABSTRACT

The hazards associated with tritium and ^{147}Pm activated luminous devices are considered. The primary hazard of tritium is due to the amount of tritium oxide which may be present. Eleven capsules were investigated for initial tritium purity and also for the amount of oxide that may be released in the event of an accidental breakage in room air. It was found that as much as 30% of the activity may be in the oxide form. The primary hazard of ^{147}Pm is that of ingestion and the resulting dose to the LLI. An examination of a reasonable hypothetical radiological accident indicated that a dose exceeding 5 times the maximum permissible intake could be received from either a tritium or a ^{147}Pm luminous capsule.

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I. INTRODUCTION+

The hazards associated with radioactive luminous devices depend, in general, on the particular isotope,^{1*} the activity per device^{2,3} and the method of fabrication. As late as 1958, according to a survey published in Nucleonics,⁴ as many as 11 installations in New York state were using radium as the activator of phosphors in luminous dials. Tritium was the only other radioactive isotope reported as being used in the New York luminising industry at that time.

During 1963 and 1964 a survey was made of the working conditions and environmental contamination in luminising establishments in the United Kingdom. Vennart,⁵ in discussing this survey, reported that seventy-five (75) people from some 23 establishments utilizing radium were measured for body radioactivity. The survey indicated that about 40% (30 people out of 75) had radium contents between 0.01 and 0.09 μ Ci (i.e. between 10% and 90% of mpbb) and two contained 0.1 μ Ci or more. He reports that the use of tritium in the luminising industry was begun about 1963, and stated that during 1963-1965 measurements of the tritium in the urine of workers frequently indicated contents greater than the mpbb (mpbb \sim 2 mCi of tritium), sometimes two or three times the mpbb. He also reported that at least two factories abandoned tritium and returned to using radium because of the objections of the workers to submitting urine samples. Vennart presented a strong case for the continued use of radium because of the relative ease of biological monitoring.

The problems associated with radium activated luminous devices are well known.⁶ The U.S. Army has essentially banned further use of luminous devices activated by radium paints, mainly because of the problems of leakage and contamination and radiation damage to the phosphor at high luminosity levels. Major problems have also been

⁺ Portions of this report were presented at the 1968 Health Physics Society Annual Meeting, Denver, Colorado, June 18, 1968.

^{*} Superscript numbers denote references which may be found on page 37.

encountered in the use of tritiated luminous paints; experience has shown that, in general, tritium is not incorporated in a chemically inert form in optically useful plastics such as polystyrene.⁵ At the present, great emphasis is being given to the use of tritium in the gaseous form as an activator of phosphors in heat-sealed glass type luminous devices. In addition to gaseous tritium, one now has a choice of at least a half dozen different radioactive isotopes as the activator for self-luminous devices and at least as many different variations in the method of fabrication. This present investigation is concerned only with evaluating the hazards presented by tritium (gaseous) and promethium (^{147}Pm) as phosphor activators. Further, our main concern is for the user personnel, rather than for the luminising workers.

Luminous capsules manufactured by different processes have been evaluated for potential inhalation and skin absorption hazards in the case of tritium, and ingestion hazards in the case of ^{147}Pm . A photograph of some typical devices is shown in Figure 1. Although the tritium gas, when sealed in a capsule, may be of high purity, oxidation may occur between the gas and the phosphor or other impurities in the capsule. Quantitative evaluation of the amount of tritium oxide present is essential. Osborne⁷ has shown that tritium oxide is readily taken into the body through the lungs and skin. The oxide is considered 100 to 1000 times more hazardous than the gaseous $^3\text{H}_2$.⁸ In addition to the HTO (H^3HO) which initially may be present inside the capsule, catalytic contaminants may be present external to the capsule which could, in the event of accidental breakage, lead to a significant conversion of tritium gas to HTO under "normal" atmospheric conditions.

Eleven tritium gas activated luminous capsules (glass) were broken in a reaction test chamber and the reaction products analyzed for tritium oxide. Test chamber conditions during 5 of the reactions simulated the conditions one might encounter in the event of an accidental breakage in the field.

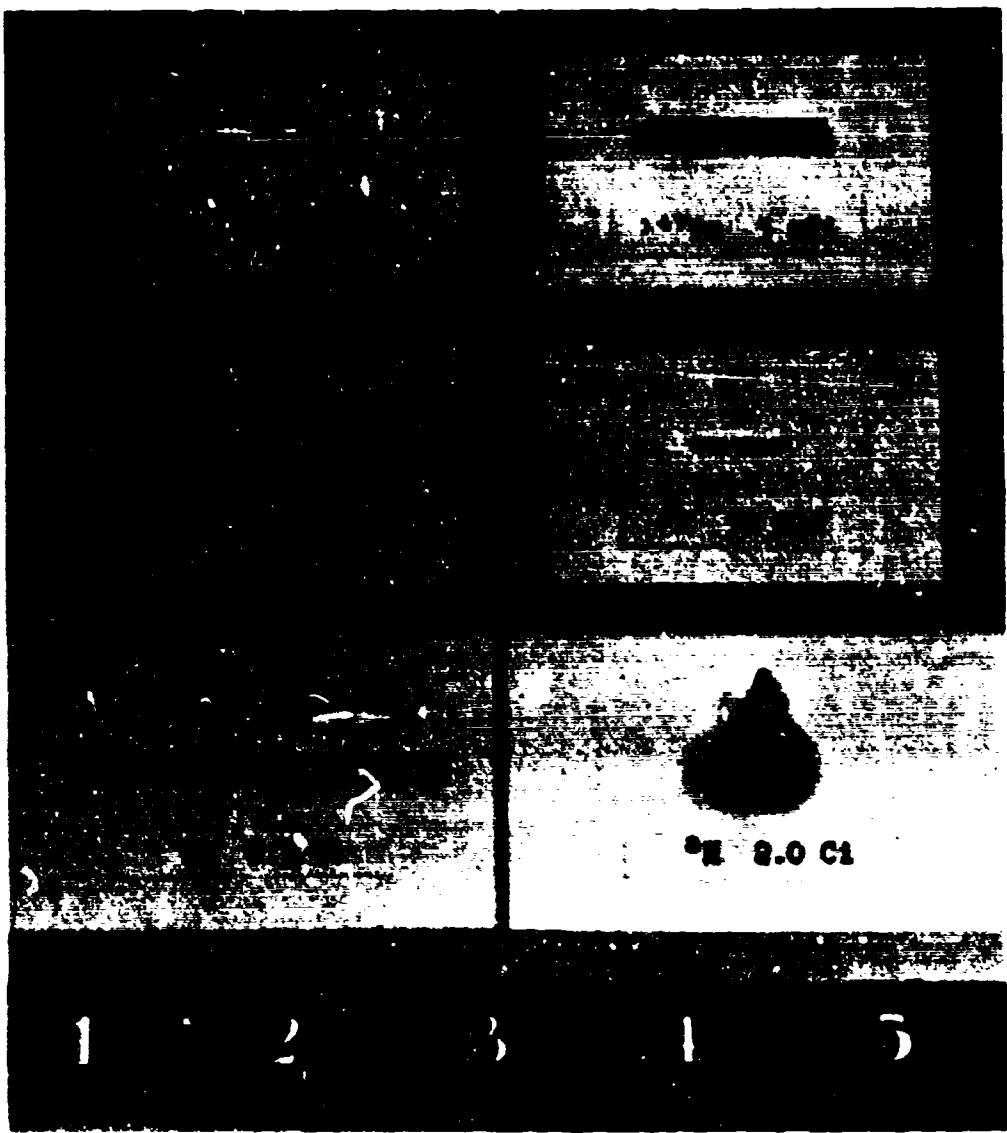


Figure 1. Photograph of Some typical Luminous Devices

The relative ingestion hazards between ^{147}Pm radioactive luminous devices manufactured by two different processes were also investigated. In this case the investigation simulated the maximum hazard which might occur if the device were crushed and ingested.

Routine leakage studies were also made on 172 luminous devices, representing both ^3H and ^{147}Pm . These studies were directed mainly toward evaluating the contamination problems to be expected.

II. TRITIUM

A. Discussion of Hazards

The hazards presented by the use of tritium gas in luminous glass capsules depend, very strongly in the event of tritium release, on the fraction of the total activity which must be considered as tritium oxide. The immediate presence of tritium oxide is of primary importance because of its almost complete absorption if inhaled and almost equal intake through the skin.⁷ In addition, because of its greater molecular weight, (≈ 22) the relatively heavy tritium oxide will mix quite well with air (m.w. ≈ 28) and thus will be removed only as the normal air is circulated. On the other hand, gaseous tritium would readily diffuse upward and away from user personnel under normal conditions. In fact the radiation safety personnel for operation HENRE⁹ stated that "due to its preponderant tendency to diffuse upward in free air, large releases of tritium (up to 1000 curies/hr) at any significant altitude (> 100 ft) would not give rise to ground-level concentrations above the MPC_a for tritium". The Radiological Health Handbook⁸ gives the MPC_a as $2 \times 10^{-5} \mu\text{Ci/cc}$ for $^3\text{H}_2\text{O}$ and the MPC_a as $2 \times 10^{-3} \mu\text{Ci/cc}$ for $^3\text{H}_2$.

The release of tritium at ground level, or in a building, would not necessarily result in immediate dilution due to diffusion. For example, in a study at Los Alamos¹⁰ it was found that the motion of tritium inside a closed building was controlled primarily by turbulent air currents, and was therefore quite unpredictable. For these reasons, knowledge of the fraction of the total activity which must be considered as tritium oxide is essential. In gaseous tritium

activated luminous devices fabricated from glass, tritium oxide (HTO or T_2O) may be formed by the presence of possible catalysts in the phosphor since the gas must be placed in intimate contact with the phosphor due to the very low beta energy of tritium. The oxide can also be formed very slowly by oxidation or exchange with hydrogen in water vapor if the tritium gas is released into the air by breakage of the sealed container. The luminous container for tritium must consist of a transparent material; glass appears to be the best choice. Coffin,¹¹ of Los Alamos, reported at the 11th Health Physics Society Meeting that plastic materials act as a "reactor in which tritium gas is transformed into HTO vapor". Furthermore he states that the HTO can diffuse through the plastic at least 100 times faster than the tritium gas. All of the gaseous devices reported on in this presentation contained the tritium (and phosphor, when present) heat-sealed in glass. The diffusion rates of tritium through glass have been calculated and are considered unimportant at room temperatures.

The conversion of "pure" tritium to tritium oxide due to both oxidation and exchange with water vapor has already been investigated. Dorfman,¹² using pure reactants and a mass spectrometer, has studied the reaction between HT and O_2 at relatively high tritium concentrations, and found that the initial rate was directly proportional to the tritium gas pressure with a first order reaction rate constant of $1.2 \times 10^{-4} \text{ min.}^{-1}$. This means that any given concentration of HT in O_2 will be half converted to HTO in about 96 hours. Dorfman's data are for HT and not T_2 which should be the initial form of the tritium in the lum devices in this investigation. If the beta energy effects are similar to those observed for X-rays¹³ the reaction rate for T_2 could be faster, but probably would not be greater than twice the HT value. Therefore, the half life for the tritium exchange would still be about 48 hours.

Casaletto et al¹⁴ have studied the oxidation of tritium at lower tritium concentrations and in atmospheres of O_2 and dry air. Second order reaction rate constants of $1.2 \times 10^{-3} \text{ ml/mCi hr}$ in O_2 and

0.62×10^{-3} ml/mCi hr in dry air were determined. These rate constants apply to tritium concentrations from 1×10^{-2} mCi/ml to 1 mCi/ml.

The investigation of Casaletto et al¹⁴ also included a study of the reaction rate's dependence on the O₂ partial pressure. The reaction rate was unaffected even when no oxygen was initially added. Apparently the tritium is capable of reacting with small concentrations of oxygen present as impurities. The investigation also showed an increase by a factor of approximately three in the reaction rate due to the presence of water vapor. Reactions with ordinary atmospheric air were generally the same as with dry air.

Yang and Gevantman,¹³ using pure reactants, a rather involved condensation procedure for trapping out the HTO, and a liquid scintillation spectrometer for counting the tritium, have studied the isotopic exchange between tritium and water vapor. A second order reaction rate constant of 1.5×10^{-3} ml/mCi hr was determined. Reaction half life times are shown in Figure 2 for both the oxidation rates of Casaletto et al and the exchange rate of Yang and Gevantman. Also included in Figure 2 is the first order reaction rate data of Dorfman which applies to the case of a relatively high initial tritium concentration. The continuous lines represent the regions actually investigated; the dashed lines are extrapolations.

Although the reaction half-life times are negligible for large volumes (or low tritium concentrations), such as the case in the event of the breakage of a capsule containing a few millicuries in a room, it should be noted from Figure 2 that where relatively high tritium concentrations occur and assuming the presence of an oxidizer, the conversion to HTO or T₂O can be expected.

The present investigations have been directed primarily toward a determination of the fraction of the total tritium activity which should reasonably be considered tritium oxide in the event of accidental breakage of the glass capsule in ordinary air. The activity per capsule may range from 10^{-3} to 10 Curies. A single piece of military equipment may contain numerous capsules. 14

1. TRITIUM EXCHANGE WITH WATER VAPOR (YANG AND BEVANTMAN).
2. TRITIUM OXIDATION IN OXYGEN (CASALETTO, ET AL).
3. TRITIUM OXIDATION IN DRY AIR (CASALETTO, ET ALL)
4. TRITIUM CONCENTRATION AS EXTRAPOLATED FROM DORFMAN'S DATA.

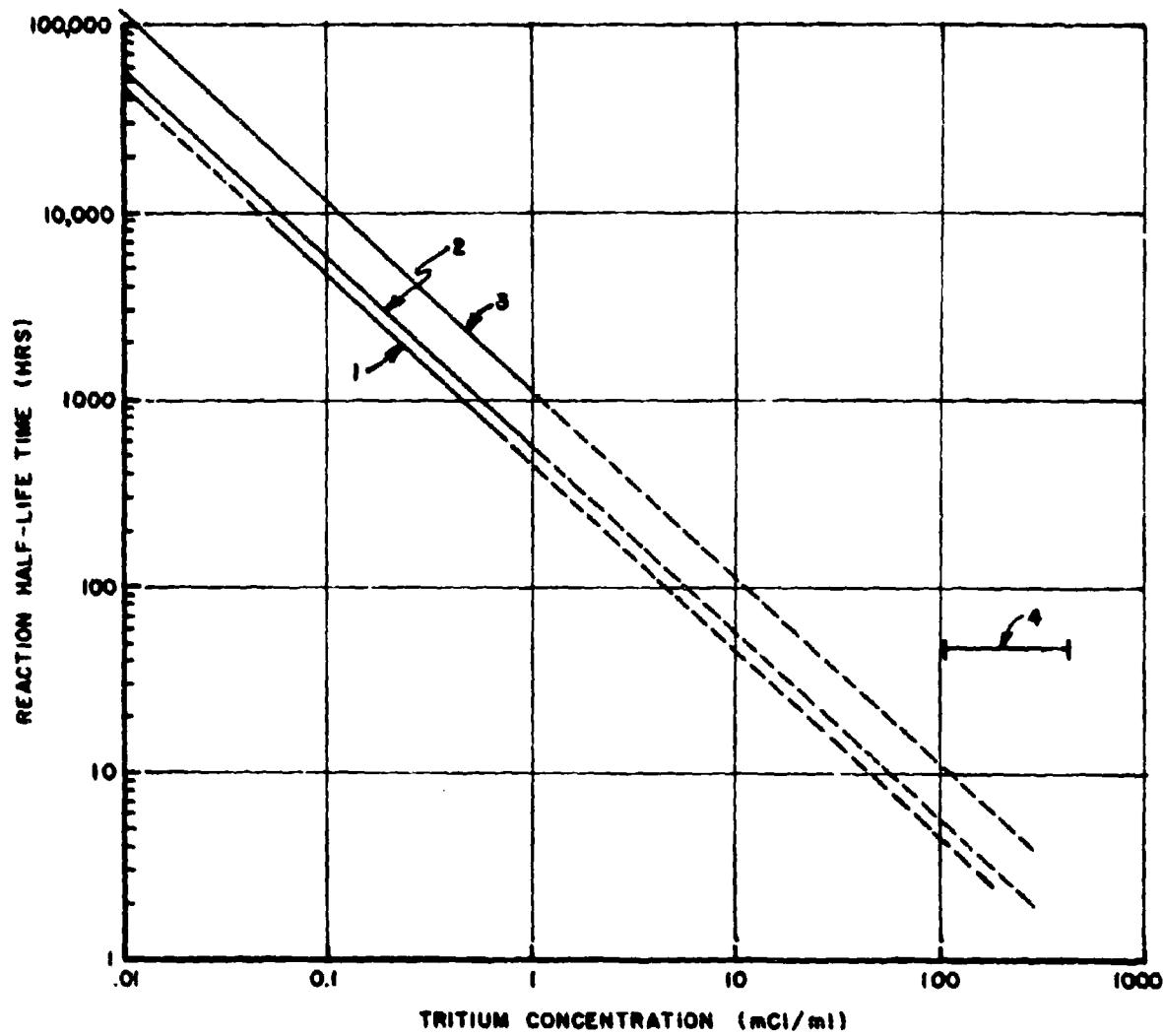


Figure 2. Reaction Half-Life Time for the Formation of Tritium Oxide.

B. Experimental Procedures

The method of collecting the H_2O or T_2O in our present investigations consisted of a simple "freeze out" system. The apparatus is shown in Figure 3 and a schematic in Figure 4. A luminous capsule was mounted on the capsule holder and positioned in the all-glass Test Chamber under controlled ambient conditions. Possible contaminants in the Test Chamber (≈ 700 cc) included rubber "O" ring seals used around the hammer and capsule holder, silicone grease used for maintaining the vacuum, epoxy used for positioning the "O" rings, and in some cases a cellulose nitrate-base cement for holding the capsule. The luminous capsule was broken by striking the outside end of the movable hammer which in turn strikes and breaks the capsule.

Prior to breaking the capsule, the system, excluding the Test Chamber, was evacuated to about 10^{-2} torr by a fore pump; a glass bead trap was placed between the fore pump and the system. The trap was cooled to liquid nitrogen temperature and served as a collector of back-diffusing hydrocarbon vapors from the pump, thus keeping these vapors from contacting the tritium system. Capsules were broken in the presence of room air and "dry" nitrogen. After the desired reaction time (generally 5 minutes), Stopcock V2 was opened allowing the reaction products to flow through Trap 1 at room temperature, Trap 2 at liquid nitrogen temperature, and into the Ballast (3×10^4 cc). Both Traps were filled with glass beads to aid in the trapping process. Trap 1 at room temperature served as a control, i.e., as an indication of system cleanliness; surface contamination due to greases would readily adsorb tritium.

Stopcock V2 was left open for 3 minutes to attain equilibrium conditions. At the end of 3 minutes V6 was opened to admit N_2 to bring Traps 1 and 2 to within a few millimeters of atmospheric pressure. Valve V2 was then closed, Traps 1 and 2 removed, and all room temperature components sealed with flexible tubing pinch-off clamps. Trap 2 was sealed at one end; the other end was connected to a tube which ran through a column of about 24 cm of liquid scintillator¹⁵ (POPOP). The

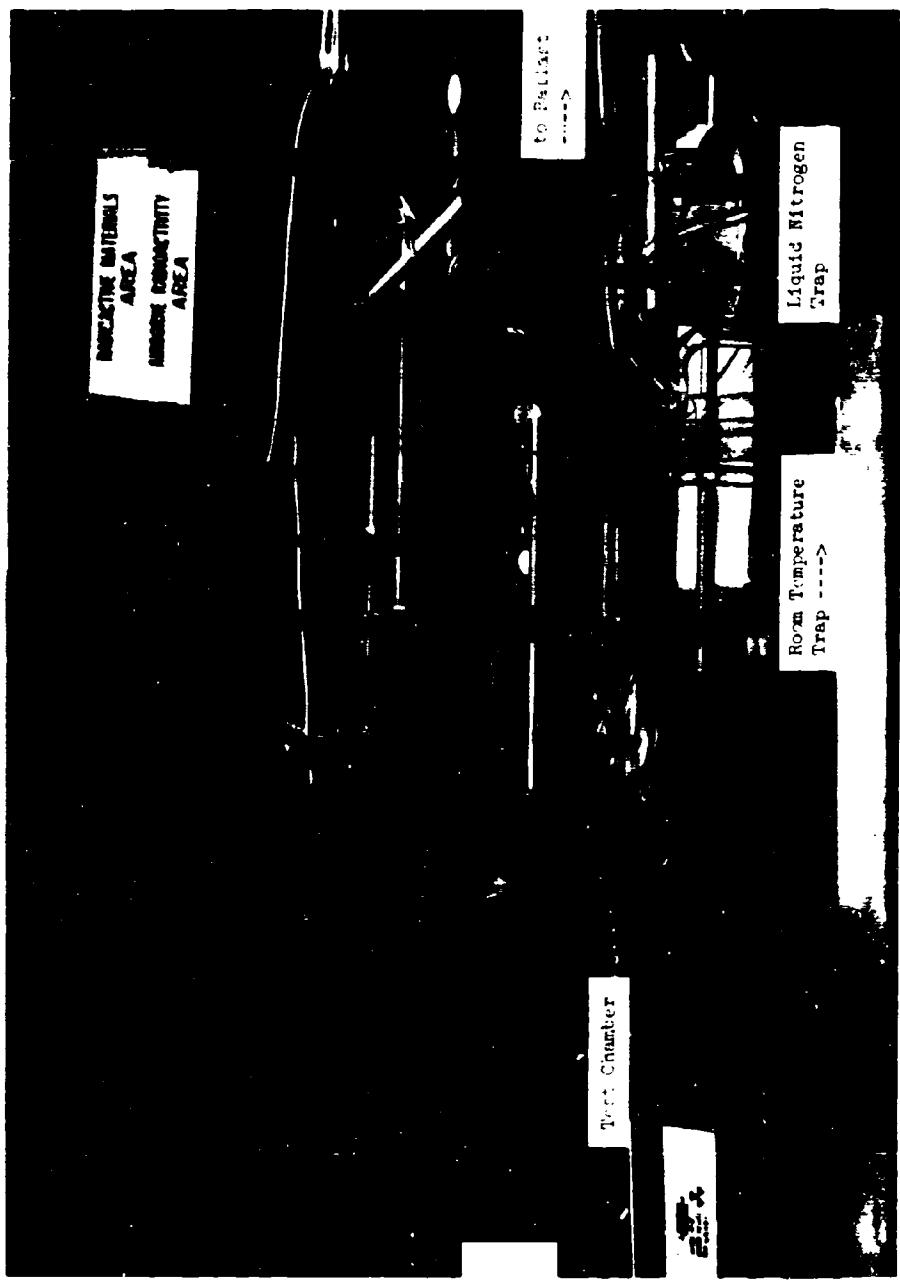


Figure 3. Photograph of the Tritium Oxide Adsorption Apparatus

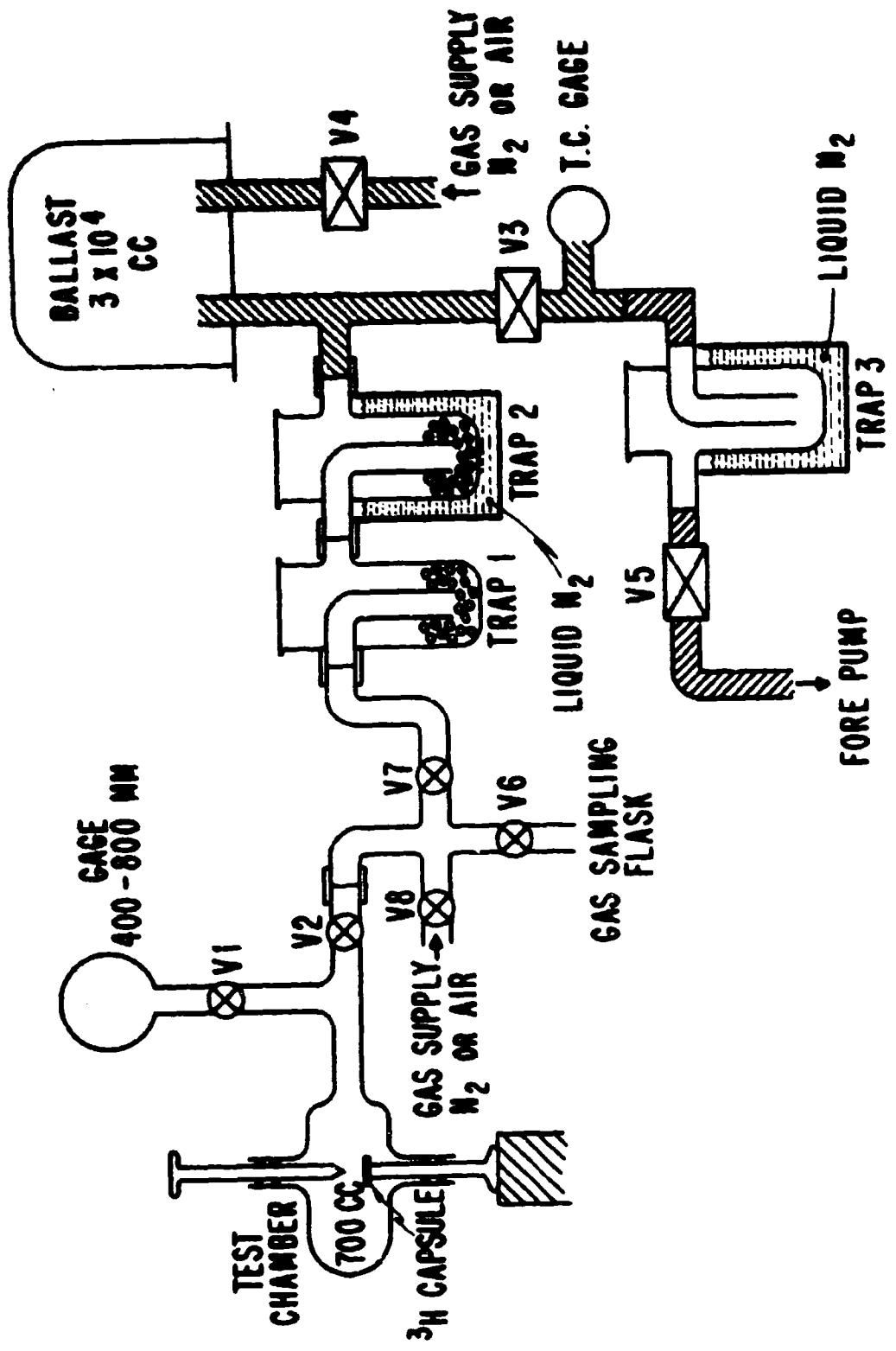


Figure 4. Tritium Oxide Adsorption Apparatus.

expanding gas from Trap 2, as it warmed back to room temperature, was then permitted to bubble through the POPOP.

The remaining activity in the Traps was measured by first "wetting" the beads with 10 ml of ethanol followed with a rinse of 10 ml of POPOP. The ethanol and POPOP were then poured into a vial labeled Rinse 1. At least two more additional rinses were made with POPOP. The activity in the vials was diluted as necessary and then counted in a calibrated liquid scintillation spectrometer.

The tritium which reached the Ballast, as well as the tritium remaining in the Test Chamber, was measured by flushing the tritium out through a Tritium Air Monitor. It was found that the rate at which the tritium was removed was proportional to the amount present. A plot of the tritium concentration vs time is shown in Figure 5 for one of the tests. The total activity was found by extrapolating to zero time and then integrating the exponential function from zero to infinity. The tritium remaining in the broken capsule section was also measured by placing the broken sections in a vial containing POPOP.

Typical data from all the rinses of the cold and warm Traps, the Ballast flush, the Test Chamber flush and the Capsule residue have been tabulated as shown in Table I. Shown in Table II, is a summary of the results. Luminous capsules containing gaseous tritium were broken in atmospheres of both room air and nitrogen.

C. Results and Discussion

To estimate the hazards from gaseous tritium devices we assume that the activity found in the liquid nitrogen trap (see Table II) is due to the "freezing-out" of tritium oxide. To substantiate this assumption we have evaluated the effect of temperature on the adsorption of tritium. In this temperature dependence study three traps were used in series and at different temperatures. The reaction products were allowed to pass through first, a trap at room air temperature, second, a trap at a solid CO_2 - acetone mixture temperature, and third, a trap at liquid nitrogen temperature.

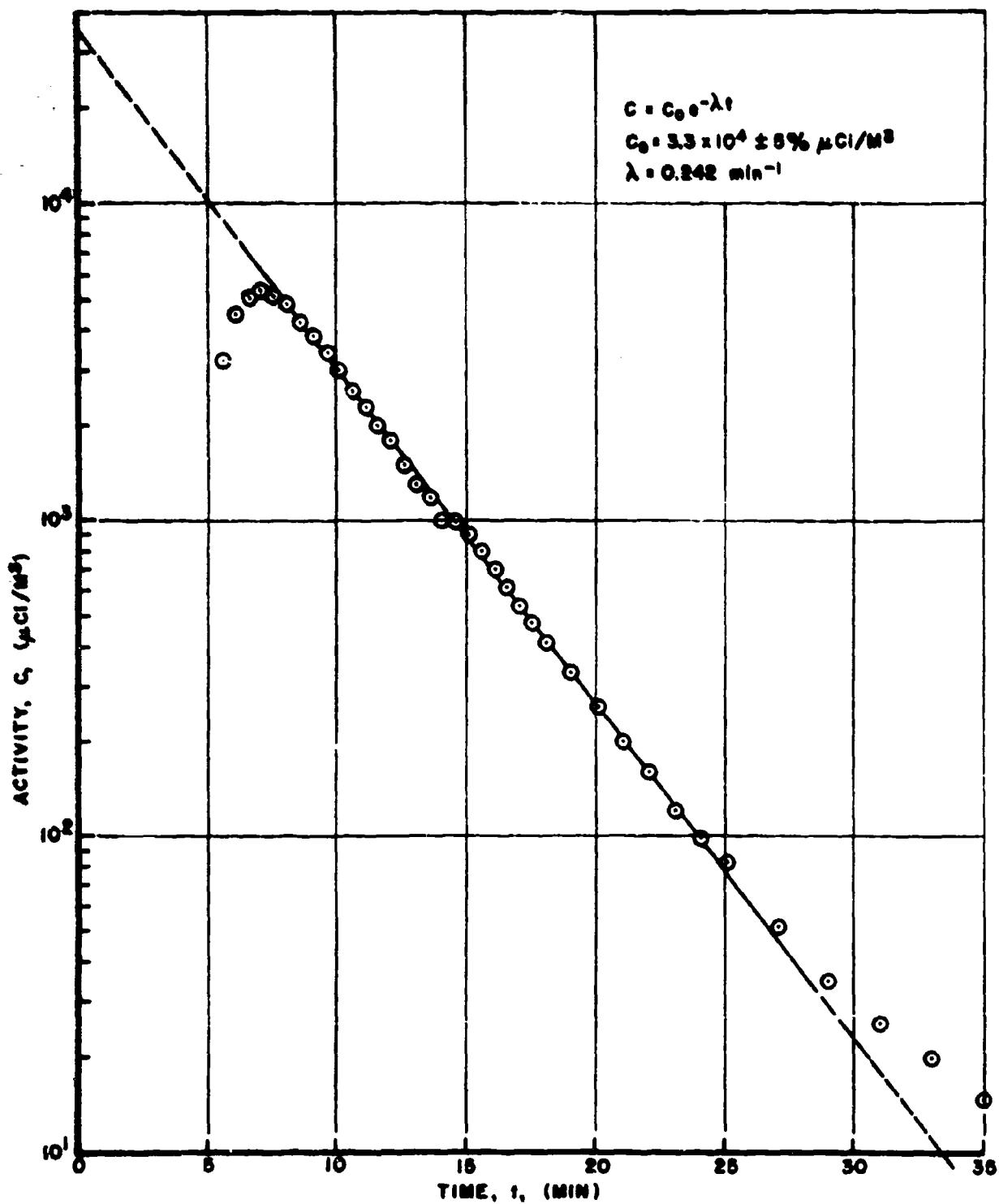


Figure 5. Ballast Flush (Test 5)

Table I. Summary of Activity Recovered from Typtrol Tests

System Conditions	Test 6	Test 7	Test 10 (Calibration)	Test 8	Test 9
Test Chamber Atmosphere	room air	room air	room air	R_2	R_2
Water Vapor Density (mg/l)	7.6	10.3	10.4	0.003(R_2)	0.001 to 0.003
Total Pressure (mm of Hg)	780	761	760	692	692
Cold Trap Condition	clean	clean	clean	6.9 μ g H_2O pre-cast	clean
21					
Cold Trap Rinse 1	1000 μ Ci	1083 μ Ci	1095 μ Ci	481 μ Ci	597 μ Ci
Rinse 2	169	290	197	86	82
Rinse 3	30	79	64	16	16
Rinse 4	6	16			
Rinse Total	1295	1468	1356	583	695
Estimated Total	1305	1475	1390	530	700
Warm Trap					
Rinse 1	13.00	0.32	1.32	20.9	35.8
Rinse 2	2.05	.45	.23	3.6	5.8
Rinse 3	.56	.16	.06	.9	1.8
Rinse 4	.28	.13			
Rinse Total	15.89	1.06	1.61	25.4	43.4
Estimated Total	16.2	1.50	1.66	26.0	44.5
Ratio, Cold Total to Hot Trap Total					
Cold Trap Expansion	80.5	1030	840	22.7	15.7
Ballast Flush	640-2240	2200-2700	7310-8930	1760-2380	1500-1840
Test Chamber Flush	450-830	290-420	0.11-0.55	500-920	450-840
Broken Capsule Residue	176	206	160	315	122
Total	$3.58 \pm 2.0\%$ μ Ci	$4.48 \pm 8\%$ μ Ci	$9.67 \pm 9\%$ μ Ci	$3.71 \pm 1.6\%$ μ Ci	$3.18 \pm 1.2\%$ μ Ci
Initial Amount (mCi)	$13.0 \pm 2.0\%$ μ Ci	$13.0 \pm 2.0\%$ μ Ci	$10.0 \pm 10\%$ μ Ci	$13.0 \pm 2.0\%$ μ Ci	$13.0 \pm 2.0\%$ μ Ci
Percentage Recovered	17.44%	27.47%	14.21%	20.41%	18.34%

Table II. Summary of Tritium Capsule Breakage Tests

Initial Purity	atm	Water Vapor Density mg/l	Percentage of Activity Measured in Cold Trap to Activity Entering Trap	
			Cold Trap Surface Condition	Test
Test 3	N_2	0.008 (max)	clean	10 - 14%
Test 9	N_2	0.001 to 0.003	clean	28 - 32
Catalytic Surface Reaction				
Test 3	N_2	0.008 (max)	6.9 mg H_2O pre coat	20 - 25
Room Air (5 runs)	Air	6.7 to 14.7	clean	29 - 67
System Calibration (Oak Ridge Capsule)				
Test 10	Air	10.4	clean	13 - 16
System Calibration (Oak Ridge Capsule)				
Test 11	N_2	0.001 to 0.003	clean	12 - 17

The test chamber atmosphere for each test is given under atm. The Cold Trap Surface Condition indicates the condition of the glass surface. The traps were cleaned by rinsing with xylene, conc. sulfuric acid, distilled water and ethanol. The traps were replaced after 3 or 4 tests. The glass beads were cleaned by rinsing with xylene and new beads were used for each test. For Test 8 about 6.9 mg of water vapor was transferred into Traps 1 and 2. This test was designed to enhance possible catalytic surface reaction, however no enhancement was noted.

As with the earlier tests a negligible amount of activity was trapped out in the trap at room temperature. From 7 to 8% of the activity which entered the trap at the dry ice temperature was trapped out while only about 1% was trapped out in the trap at the liquid nitrogen temperature. Although 7 to 8% of the activity trapped out at the dry ice temperature was less than that for any of the 5 runs in room air, as summarized in Table II in which only a liquid nitrogen cold trap was used, this variation might well be due to a variation among different types of tritium devices. The luminous capsule used in the dry ice run was supplied by the same company as were those used in the earlier tests but was of a different configuration and contained a different "nominal" amount of activity. Thus we feel that the results of this test show that first, adsorption of pure tritium in the liquid nitrogen traps is negligible (this is to be expected since the critical temperature for tritium is probably within a few degrees of the critical temperature of hydrogen which is 33°K), and second, the possibility of a weak van der Waals' catalytic reaction caused by adsorbed surface impurities is also probably of minor concern, again, as indicated by the relatively small amount of activity recovered from the liquid nitrogen trap. We therefore conclude on the basis of all tests made to date, that when a luminous capsule containing tritium is broken, particularly in room air, a sizable fraction of the total activity should be considered to be in the form of tritium oxide.

Unexplained in Table II is the rather large amount of activity trapped out in the two tests with the capsules obtained from the Oak Ridge National Laboratory. The radioactive purity of these capsules was stated to be initially greater than 99%. If our system actually converts some tritium to the oxide form, then the amount trapped out from the Oak Ridge Capsules represents an "upper limit" correction to the amount of tritium oxide recovered in the cold trap for the tests with the luminous capsules. Taking this in to account and averaging the activity recovered from the cold traps from the room air tests with the luminous capsules, we still find that at least 25-30% of the

total activity released should be considered as tritium oxide.

The initial purity at the time of breakage of the luminous capsules can also be estimated following the above procedure for those capsules broken in a nitrogen atmosphere. For the three capsules tested, we find that the amount of tritium oxide initially present may range from a negligible amount, i.e. about the same as the Oak Ridge capsules, to as much as 19% (Test 9).

Tests to date have been limited to capsules containing only small amounts (~ 10 mCi) of tritium. Extrapolating our present data to the curie level tritium capsule, we may estimate the maximum radiological hazard to be expected on the basis of the values given by Lawrence et al³ for the maximum permissible intake (MPI) for a single acute exposure. Assuming that the breakage of a curie capsule releases about 100 mCi of tritium oxide in a small volume of several cubic meters ($\sim 2 \times 10^3$ liters), such as in a small military vehicle, we may use Osborne's⁷ results to calculate the dose. Using an average value from his results for unprotected subjects of $15 \mu\text{Ci}/\text{min}$ per $\mu\text{Ci}/\text{l}$, we obtain an intake rate of about $750 \mu\text{Ci}/\text{min}$. Of course, this would not continue for very long since our initial average concentration would be only about $50 \mu\text{Ci}/\text{l}$. In several hours it might be expected that our subject might assimilate a maximum of some 50 mCi of the total 1 curie released. Although this value appears quite high, Donth and Maushart² have reported an intake of up to 10% of total tritium activity in an incident at the Karlsruhe Nuclear Research Center involving the production of luminous paint. According to Lawrence et al³ a single acute intake of 15 mCi of tritium will give a dose of 5 rem in one year. Brodsky and Beard¹⁶ have estimated that a single intake of about 50 mCi of tritium oxide will give about 5 rem to the whole body within one year. Thus, on the basis of Lawrence's study and our present results, we might anticipate maximum exposures of 10 rem or more from accidents involving multicurie gaseous tritium luminous devices in confined areas.

III. PROMETHIUM DEVICES

A. Introduction

The ¹⁴⁷Pm luminous devices which were studied were fabricated by two different methods. Type I consisted of ¹⁴⁷Pm imbedded in ceramic microspheres, mixed with a phosphor, and contained in a glass capsule. Type II consisted of ¹⁴⁷Pm and a phosphor incorporated in a plastic rod. The primary purpose of the present study was to estimate the relative potential assimilation hazards between the two types in the event of an accident. Wipe tests between the two types indicated no removable activity for the Type I capsule. For the Type II capsule the removable activity exceeded 0.001 μ Ci for all the rods investigated.

B. Experimental Procedures

The luminous devices were broken by two different procedures: (1) the capsule or rod was held with a pair of pliers and sheared with a second pair of pliers, one fragment of the broken capsule or rod was then crushed with a pair of long nose pliers; and (2) the entire capsule or rod was crushed with a pair of pliers to produce a larger number of smaller particles than obtained from the first procedure. In both tests the device was immersed in 100ml of distilled water while it was broken. The fragments from each of the broken capsules and rods were subjected in succession to a water immersion test, a gastric juice test,¹⁷ and a concentrated hydrochloric acid test. Test conditions, including immersion times, are shown in Table III. The capsules are immersed in the test liquid which was contained in a Buchner type disc funnel as shown in Figure 6. At the end of the immersion period the liquid was pulled through the funnel by an aspirator. The nominal maximum pore size diameter of the funnel, as given by Corning Glass Works, is 0.9 to 1.4 μ . The soak with the test liquid was followed by at least two rinses to reduce the effects of cross contamination between successive liquids. A new funnel was used for each fracture test. The test liquid and rinses were sampled for activity by evaporating a known amount on a planchett and measuring the activity with a wide β type instrument.

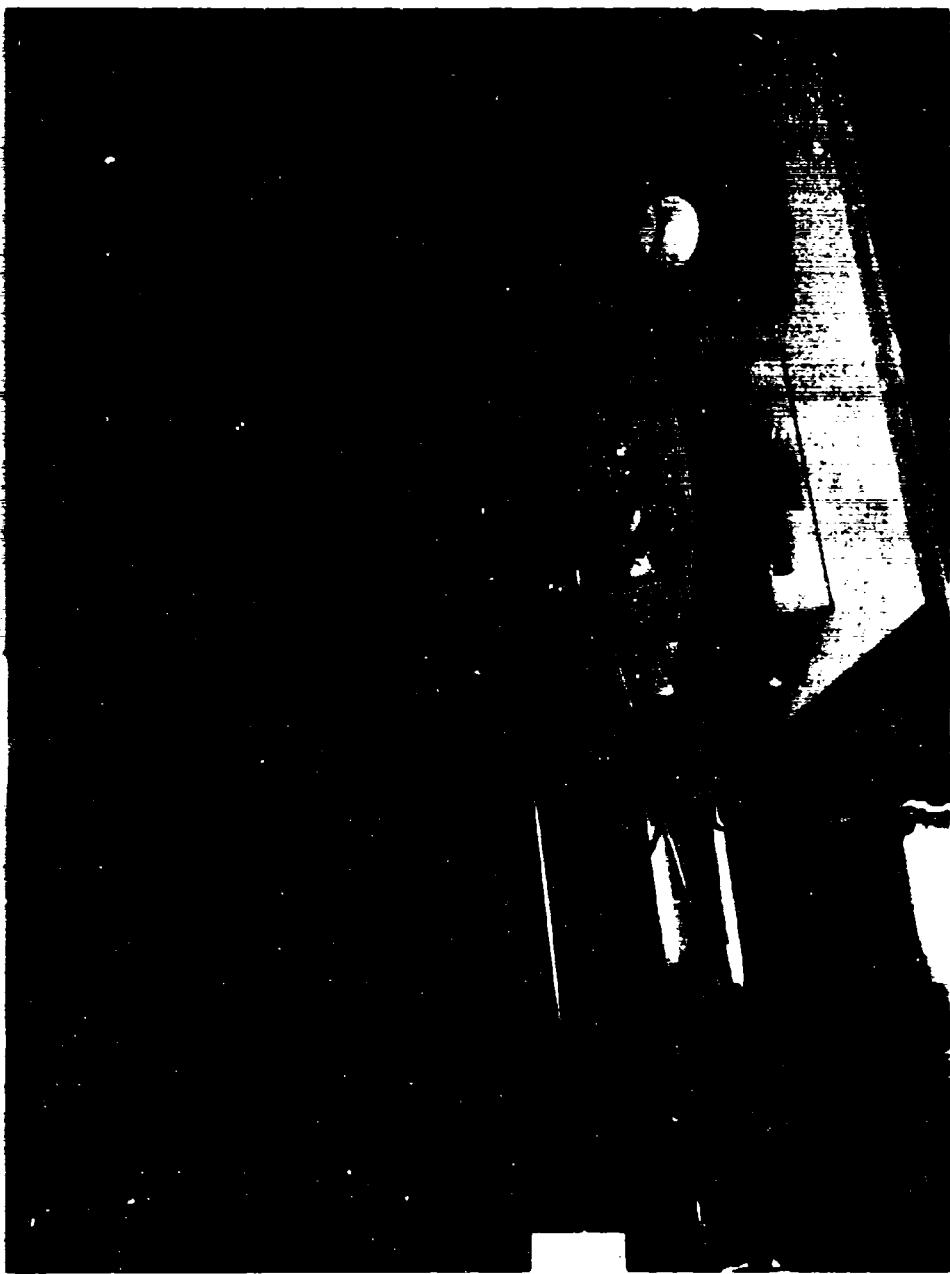


Figure 6. Photograph of ^{147}Pa Leach Test Set-up

Table III. Estimated Activity Leached From Broken ^{147}Pm Radioactivated Luminous Devices

Liquid	Soak Time (hrs)	Stir Time (hrs)	First Test			Second Test			First Test			Second Test		
			Microsphere Capsule 0.68 μCi			Plastic Rod 1.5 μCi			First Test			Second Test		
			Activity Leached (μCi)	% Leached	% Assimilated	Activity Leached (μCi)	% Leached	% Assimilated	Activity Leached (μCi)	% Leached	% Assimilated	Activity Leached (μCi)	% Leached	% Assimilated
Distilled H_2O	22	6 1/2	0.6	0.09	2	0.3	0.3	0.02	0.3	0.02	0.001	0.02 ^(a)	0.001	
Gastric	2	2	6	0.9	9 $\times 10^{-6}$	40	6	6 $\times 10^{-4}$	100	7	7 $\times 10^{-4}$	310 ^(a)	21	2.1 $\times 10^{-3}$
Conc. HCl	1	1	20	2.9	40	5.9	240	16	120 ^(a)	8				

(1) Percentage assimilated based on the fraction (10^{-4}) reaching the total body from ingestion, ICPR Publ. 2 (1959).

(a) Low value probably due to a blocked filter (see text).

The estimated total activity leached by the three test liquids is given in Table III. The removal of the distilled water from the initial soak served to eliminate most of the smaller fragments from passing thru the filter during the gastric juice and concentrated hydrochloric acid tests. The second breakage test showed an increase in activity, as expected because of the additional crushing, for all test conditions for the device with microspheres. The distilled water test and the concentrated hydrochloric acid test for the Type II rods with the ^{147}Pm and phosphor in plastic showed an unexpected decrease. In the plastic device, during the distilled water test, a yellow substance formed on the unfiltered side of the funnel; conceivably this substance, possibly the phosphor, may have inhibited the ^{147}Pm from passing through the funnel. Blocking of the filter also occurred during the acid test.

The microspheres were examined for possible fracture and particle size. The top surface of the Buchner filter used for the Type I leach test was swabbed with a glass slide coated with optical grade oil. The slide was mounted on a microscope and examined for round objects. Figure 7 shows two round objects and two or three curved segments suggesting they were broken from a round object. If the spheres were broken, there probably were broken particles sufficiently small to pass through the filter. However, these smaller particles would have been eliminated after the initial rinses with distilled water and would not have interfered with the activity leached from the gastric juice test. Figure 7 shows a second picture of one isolated round object. The approximate diameter of the round objects for both pictures is 30μ .

C. Results and Discussion

The amount of activity leached from the fractured devices represents an indication of the maximum level of ^{147}Pm which is available for assimilation by the body following fracture, such as grinding, chewing, etc., and subsequent swallowing. The fraction assimilated into the total body, according to ICRP Publ. 2, 18 is 10^{-4} . Therefore, to estimate the percentage of the original capsule activity assimilated

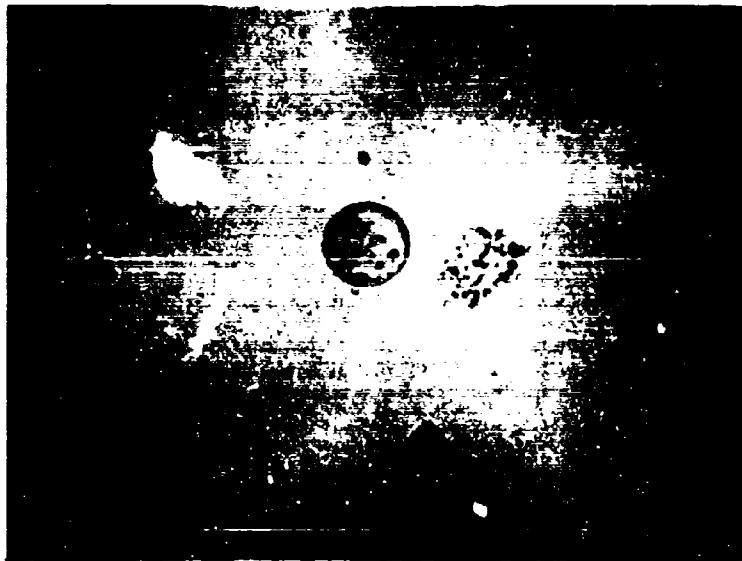


Figure 7. Round Objects Photographed from a Swab of the Buchner Funnel Containing Microspheres. The approximate diameter in both pictures is 30 μ .

into the body, the percentage leached by the gastric juice as shown in Table III should be multiplied by 10^{-4} . The adjusted percentage assimilated values are also shown in Table III.

An additional indication of the amount of ^{147}Pm which might be assimilated can be obtained from an extrapolation of studies performed with rats at the Southern Research Institute¹⁹. Rats were force fed microspheres encapsulated in gelatin. The microspheres are similar to the Type I capsules described above. The entire capsule reached the stomach where the gelatin rapidly dissolved. The animals were sacrificed at various times as shown in Table IV and examined for activity. The alimentary tract was intentionally excluded from the carcass. Activity found in the carcass and urine represent a measure of the activity actually absorbed in the body, even if only temporarily as in the case with urine, while most of the residue activity remaining in the alimentary tract should still be eliminated. This was the case with the two animals sacrificed at 96 hours where the activity found in the alimentary tract was only 5 - 7 % of that found in the carcass plus urine. Animal No. 4 revealed an activity substantially higher than the other animals which may have been due to slight hemorrhaging immediately after being force fed the capsule.

It is assumed, following the SRI report,¹⁹ that man would absorb about the same percentage of ^{147}Pm as would a rat. Although such an estimation is admittedly crude, the results are of interest. Given in Table IV are the percentages of original activity which were obtained in the carcass plus urine. These figures, considering Animal No. 4 as abnormal, represent a rough order of magnitude agreement with the percentage assimilated values given under Type I in Table III.

Since the fracture tests undoubtedly involved broken microspheres as revealed in Figure 7, a lower absorbed activity from the rat studies would appear reasonable. A comparison of data, however, between the two sets of experiments does not support this hypothesis.

To evaluate the assimilation hazard of ^{147}Pm , assume that a 300 mCi

Table IV. Activity Data Taken From Southern Research Institute Report (1) On Microspheres With Data Reduced for Comparison With Activity Leached From the Broken Fm-147 Radioactivated Luminous Devices

	Animal No. 1	Animal No. 2	Animal No. 3	Animal No. 4	Animal No. 5	Animal No. 6
Sacrifice Time (hours)	48	48	72	72	96	96
cps in Microspheres Administered ($\times 10^6$)	0.88	1.18	1.03	1.03	1.06	0.86
Carcass (cps)	30.5	5.09	1018	38,500	30.9	971
Urine (cps)	636			9.26	853	3.1
Sum of Urine + Carcass (cps)	666	5.09	1018	38,509	884	974
% of Original Activity Obtained in Carcass + Urine	7.6×10^{-4}	4.3×10^{-6}	9.9×10^{-4}	3.7×10^{-2}	8.3×10^{-4}	1.1×10^{-3}

(1) Animal Studies Related to Possible Radiation Hazards of Plutonium-147 Contained in Microspheres, Southern Research Institute, Birmingham, Alabama (Sept., 1961).

^{147}Pm luminous source of the type now available is completely crushed and 10% is ingested, admittedly a rather remote possibility. The amount of activity assimilated based on the above results would represent an average of $7 \times 10^{-4}\%$ of the ingested activity, or about $0.2 \mu\text{Ci}$. From the ICRP Publ. 2,¹⁸ about 35% of this amount would end up in the critical organ, the bone. Since the maximum body burden for ^{147}Pm in the bone is $69 \mu\text{Ci}$,²⁰ then the activity in the bone would be of no biological consequence.

Of more concern in the present ingestion studies is the dose equivalent to the G. I. tract based on a single ingestion. From the model for the G. I. tract,²¹ a single ingested intake of about $600 \mu\text{Ci}$ would deliver 7-1/2 rem to the critical organ, the lower large intestine. This value for an MPI compares with a calculated value taken from Brodsky and Beard¹⁶ of $580 \mu\text{Ci}$ in the G. I. tract to give 5 rem in 1 year. Tracerlab²² has also examined the dose to the lower large intestine due to the passage of one $0.1 \mu\text{Ci}$ ^{147}Pm microsphere. A dose of 0.88 millirem per $0.1 \mu\text{Ci}$ of ^{147}Pm was calculated. The $600 \mu\text{Ci}$ MPI estimated in the present study implies, based on the Tracerlab study, a total dose to the lower large intestine of about 5.3 rem. These small differences in the calculated doses are not important.

The accident considered above involving the ingestion of 10% of a 300 mCi luminous source would therefore exceed the MPI by a factor of 50. It is therefore conceivable that the MPI (for the dose to the G. I. tract) could well be exceeded due to the partial ingestion of either of the two types of ^{147}Pm devices considered.

When inhalation is present, which is probably not important for the Type I microspheres because of their size, then the fraction accumulated in the critical organ (bone) is given by Brodsky and Beard²³ as 9%. The activity required to give 5 rem to the critical organ within one year, and based on inhaling soluble material, is only $181 \mu\text{Ci}$.¹⁶ The dose to the lungs should also be investigated if inhalation is suspected. As an illustration, consider the ^{147}Pm particles to be

insoluble and to vary in diameter from 1.0 microns to 50 microns. For 1 micron diameter particles about 25% would be expected to end up in pulmonary regions of the lung, while at 50 microns the amount deposited would have dropped to about 3%.²⁴ The required insoluble activity inhaled to give a dose of 5 rem per year to the lung is about $80 \mu\text{Ci}^{16}$. Assuming only 10% is deposited in the pulmonary regions (such as the case for a 20 micron diameter particle), then 800 μCi would have to be inhaled to give a 5 rem dose to the lung.

Due to the more easily removable surface activity of the Type II device (Section 3.1), it is obvious that the Type II device presents a greater potential contamination hazard, although the possibility of this removable surface activity reaching a critical organ in significant amounts is still relatively low. For example, from the handling of one Type II capsule, at least 0.005 μCi could be removed, which, of course, exceeds the limit set for removable surface activity. Of this, up to 25% could be assimilated due to inhalation while only approximately $1 \times 10^{-2}\%$ could be assimilated by ingestion.

IV. CONCLUSIONS

The maximum hazards to user personnel due to acute exposures to an accidental breakage of a tritium or a ^{147}Pm luminous device have been estimated in terms of maximum permissible intakes (MPI), and our evaluation of the activity available for assimilation. These hazards have been summarized in Table V. The figures given, of course, can only be regarded as rough approximations. Each potential hazard which might arise should be considered separately whenever possible. For example, the hazards due to the release of tritium will depend on the individual's position relative to the point of tritium release, convection currents, the time of exposure and the amount of tritium oxide present. For these reasons, one of the more hazardous locations for an accident would be in a small military vehicle as was described in Section 2.3. This accident assumed the breakage of a ' Ci tritium capsule, but as indicated in Table V luminous capsules containing tritium are now available with

Table V. Summary of Radiological Hazards of ^{39}Rb and ^{147}Pm Activated Luminous Devices

Isotope and Activity Per Device	Half Life	Hazards	Estimation of Device Activity Available for Assimilation	Fraction to Organ	Critical Organ	MP for Single Intake	Estimation of Maximum Probable Intake (10% of Available Activity)
^{39}Rb 1 to $10^4 \mu\text{Ci}$	12.3Y	-12 d	Inhalation and Absorption (Conversion to Rb^{+})	1.0	Body Tissue	$15\mu\text{Ci}$ (a)	$100 \mu\text{Ci}$ (.6 MP)
^{147}Pm 10^{-1} to $300 \mu\text{Ci}$	2.7Y	-1 d	Ingestion (insol.)	1.0	III	$600\mu\text{Ci}$ (b)	$3 \mu\text{Ci}$ (.5 MP)
^{39}Rb		-1 d	Ingestion (sol.) (1 to 20% actually leached)	4×10^{-6} (c)	Bone	$1 \mu\text{Ci}$ (d)	$0.6 \mu\text{Ci}$ (.6 $\times 10^{-4}$ MP)
				$10^{-3} \mu\text{Ci}$ (f)	Lung	$80\mu\text{Ci}$ (r)	$3 \mu\text{Ci}$ (.4 $\times 10^{-3}$ MP)
				$10^{-2} \mu\text{Ci}$ (f)	1×10^{-2} (g)	$180\mu\text{Ci}$ (r)	$3 \mu\text{Ci}$ (.2 $\times 10^{-2}$ MP)

(a) Ref. 3
 (b) Ref. 16 Inhalation of insoluble activity figure multiplied by 62.5%
 (c) Ref. 18
 (d) Ref. 16 Amount injected into critical organ (48 μCi) divided by amount expected to reach critical organ (Ref. 18)
 (e) Ref. 24
 (f) Ref. 16
 (g) Based on 10% deposited in lung (Ref. 24) times 25% assimilated into body fluids directly from lung times 35% (Ref. 18) assimilated into bone.

activities up to 10 Ci. The accidental release of activity from this larger source in a confined area, of course, would lead to a correspondingly greater dose.

It is found that the amount of tritium oxide released from the breakage of tritium-activated luminous capsules in room air can be as high as 30%. This result should not be considered typical of all tritium filled luminous capsules. Only one type was investigated in the present report. Conceivably, tritium activated luminous capsules originating from different fabrication procedures might result in a smaller hazard. Also, with respect to confirmation studies, it would be highly desirable to have an independent and more direct experimental determination of the amount of tritium oxide released.

Also shown in Table V is a summary of the hazards concerning ingestion and inhalation of ^{147}Pm . Of most importance from the standpoint of an accident would be the ingestion of a capsule and the resulting dose to the LLI. Although actual ingestion may appear as a rather remote possibility, past experience, along with a wide distribution of sources, points to an ever increasing likelihood that a capsule will find its way into the food supply.

Minimizing potential radiological hazards and contamination problems is always emphasized from the health physicist's viewpoint. Thus, the efficiency of the light output per maximum hazard for the intended application should be used as the ultimate criteria for selecting a particular device. Other selection criteria of lesser importance include the sources' surface contamination, physical integrity or strength characteristics, and uniformity between sources. The last two factors, involving the fields of mechanical and quality control engineering, and although not the immediate concern of the health physicist, should nevertheless be examined in detail. The basis for a radiological accident would certainly exist during the insertion of an irregularly shaped capsule into a recessed receptacle. Among the smaller tritium activated luminous capsules, numerous irregularly shaped

capsules have been observed where apparently quality control difficulties have been encountered in forming a well-rounded uniformly shaped sealing end.

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13. ABSTRACT The hazards associated with tritium and ^{147}Pm activated luminous devices are considered. The primary hazard of tritium is due to the amount of tritium oxide which may be present. Eleven capsules were investigated for initial tritium purity and also for the amount of oxide that may be released in the event of an accidental breakage in room air. It was found that as much as 30% of the activity may be in the oxide form. The primary hazard of ^{147}Pm is that of ingestion and the resulting dose to the LLI. An examination of a reasonable hypothetical radiological accident indicated that a dose exceeding 5 times the maximum permissible intake could be received from either a tritium or a ^{147}Pm luminous capsule.		

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